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REQUIREMENTS FOR CONTROLLING A REPOSITORY'S RELEASES OF CARBON-14 DIOXIDE; THE HIGH COSTS AND NEGLIGIBLE BENEFITS

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ABSTRACT

A repository excavated within the unsaturated zone may release carbon(C)-14 dioxide in amounts that exceed limits imposed by the Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission (NRC). The release would not threaten the general population, but may expose some hypothetical maximally exposed individual to 0.0005 millirems/year (mrem/yr). Yet a repository's releases of C-14 dioxide are strictly regulated, perhaps unintentionally. The EPA and NRC regulations could force the Department of Energy (DOE) to design and fabricate an expensive 10,000-year waste package solely for the sake of controlling releases of C-14 dioxide. This paper argues that the repository regulations should exempt releases of C-14 dioxide or at least impose more equitable limits.

INTRODUCTION

Regulations written by the Environmental Protection Agency (40 CFR Part 191)[1] and the Nuclear Regulatory Commission (10 CFR Part 60)[2] limit releases of high-level radioactive waste (waste) from a geologic repository. The EPA requires that for 10,000 years, cumulative releases of specified radionuclides not exceed certain limits. The NRC additionally requires that the waste be contained substantially completely for 1,000 years, and thereafter released at an annual rate that does not exceed other limits. The EPA limits apply at the boundary of the accessible environment. For gaseous releases, this boundary would coincide with the surface directly above the repository and for aqueous releases, the lithosphere situated about five kilometers from the repository. The NRC release limits, however, apply within the repository (i.e. at the boundary between the host rock and the engineered barrier system).

At the time these requirements were written, saturated sites were the leading contenders for a repository. Consequently, the EPA and NRC limits were not intended to control gases that would be released into and through unsaturated rock. When applied to gases, these limits become overly conservative and place excessive demands on the waste package. This paper provides an argument for why the EPA and NRC release requirements should not apply to radioactive gasses, notably, carbon-14 dioxide.

Sources of Gaseous Carbon-14

Carbon-14 (C-14) is produced in the nuclear fuel element during the reactor operation primarily by neutron reactions with N-14, C-13, and 0-17 in the fuel via ^{14}N (n,p) ^{14}C, ^{13}C (n, γ) ^{14}C, and ^{17}O (n, α) ^{14}C. Carbon-14 produced by fission is negligibly small. Most of the C-14 resides in the UO_2 fuel matrix, zircaloy cladding and hardware associated with the fuel assembly, such as spacers, end fittings and boiling water reactor (BWR) fuel channels. Some C-14, however, migrates to grain boundaries and the pellet-cladding gap, thereby becoming more readily releasable upon breach of the cladding. A small amount of C-14 is also present in the crud deposit on the cladding surface (See Figure 1 for the distribution of C-14). Nitrogen impurities in the fuel is one source of C-14, and O-17 in the reactor coolant is another.[3] The Carbon-14 in the crud and possibly that in the cladding oxide layer is speculated to have originated from the 0-17 in the reactor coolant.

The distribution of C-14 in typical pressurized water reactors (PWR) and BWR spent fuels is shown in Figure 1. Available measured data shows considerable scatter for a single fuel assembly as well as considerable variation among the values for UO_2 or cladding for different fuel assemblies. In addition, it is not now possible nor is it likely to become possible to measure a statistically significant average of C-14 content in the spent fuel destined for the repository. Less than one third of the spent fuel to be emplaced in the repository exists today. Therefore, estimates of C-14, which are based on conservative values of the nitrogen impurities in the components of fuel are used in Table 1.[4]

The spent fuel assemblies, either intact or consolidated, will be placed inside a container filled with an inert gas. The container, in turn, will be placed inside a borehole in an unsaturated repository. In time, natural processes will corrode all the containers. Also, natural events, such as a rockfall, may puncture some containers while other containers may leak due to undetected manufacturing defects. When the containers fail, the solid waste will release radionuclides in both aqueous and gaseous forms, including C-14.

Spent fuel can release C-14 dioxide even before the cladding ruptures. The C-14 dioxide may be a product of thermal oxidation of C-14, perhaps assisted by the gamma radiation field.[5] Current data indicates that the release rate from the external surface of the fuel assembly is orders of magnitude higher than the release rate measured from the ruptured rod. Thus, the portion of C-14 inventory that can be released from the surface of fuel assemblies, presumably from the crud deposit and the oxide layer of the cladding, is termed the "rapid release fraction of C-14".[6]

Assuming that carbon-14 dioxide would be released at the same rate at which it is produced (the rate at which carbon-14 dioxide is produced increases with temperature.[7]), the Department of Energy (DOE) reported in its site characterization plan (SCP) that release rates would be highest while the waste is hot and then drop as the waste cools. The DOE reasoned that if the container could stay intact past the peak of the thermal cycle (which occurs at roughly 300 years), oxygen could not enter the container, combine with C-14 and produce C-14 dioxide.

Based on a single measured value of 0.26 percent, the DOE also reported in its SCP, that the rapid release fraction of C-14 would conservatively comprise approximately 1 percent of the entire C-14 inventory. More recent research, however, indicates that spent fuel could produce C-14 dioxide in an oxygen-free environment and rapidly release as much as 5 percent of its C-14 inventory, as C-14 dioxide.

Figure 1. Distribution of C-14 in Spent Nuclear Fuel.

Table 1. Carbon-14 Activities in Reference PWR and BWR Spent Fuel[4]

	PWR	BWR
Burnup (Megawatt Days/Metric Tons of Heavy Metal)(MWD/MTHM)	33,000	27,500
C-14 Inventory (Curies (Ci)/MTHM)		
UO_2	0.60	0.54
Zircaloy	0.35	0.76
Fuel Assembly Hardware	0.60	0.23
Total (Ci/MTHM)	1.55	1.53

After the SCP was issued, researchers discovered that C-14 dioxide may be desorbed from the spent fuel, i.e., produced without oxygen.[8] If confirmed, the research implies that C-14 dioxide forms and accumulates inside the waste container. Consequently, the longer the container stays intact, the higher the rate at which C-14 dioxide will be released once the container fails. When the container fails, releases of C-14 dioxide could be as significant during the later postclosure period when the waste package temperature is at near ambient temperature as during the early period when the temperature is significantly higher. Since the measurements to date have been made in a relatively short duration (10 hours), it is speculated that the gaseous rapid release fraction could be as high as 5 percent of the total C-14 inventory in the breached container.[9]

In addition to the gaseous release of C-14 from the exterior of the fuel, C-14 dioxide can also be released from water that infiltrates the waste containers. More than one-half percent of the spent fuel C-14 inventory may preferentially dissolve from the UO_2 matrix in 15 months.[10] The aqueous dissolution will continue at a lower rate for as long as water infiltrates the container. When this water leaves the container and enters the vadose zone, the C-14 rapidly exchanges with C-12 in aqueous bicarbonate ion and atmospheric CO_2 in the rock pores and eventually becomes a gas. Considering the potentially long groundwater travel time through the vadose zone, it is possible that a very large portion of all aqueous C-14 may escape as a gas.

Recent preliminary studies also show that underground releases of C-14 dioxide can reach the surface in less than one half-life of C-14 (5,730 years).[11,12] Therefore, if a significant portion of the waste containers and the fuel cladding fail before 10,000 years, the gaseous and aqueous releases of C-14 alone could violate the EPA regulation on the cumulative release of radionuclides (40 CFR 191.13) as well as the NRC requirement on the post-containment release for C-14 (10 CFR 60.113).

Problems with EPA and NRC Regulations

The Department of Energy (DOE) must design a waste package that meets the EPA and the NRC regulations. Driven by the need to control potential releases of gaseous carbon-14, as carbon-14 dioxide, no more than 30 out of 30,000 waste packages can fail per year for 10,000 years.[6,13] Otherwise, release limits in the EPA and NRC regulations would be violated (The regulations tolerate a much higher failure rate when applied to aqueous releases. Here, an insoluble waste form, rather than the waste package alone, can control release rates). This failure rate, however, assumes that only one percent of the C-14 inventory can be rapidly released as C-14 dioxide. Recent data, which was discussed earlier, and some German data indicate that significantly more than one percent of the C-14 inventory, specifically C-14 in the cladding, could be mobilized and then released after the cladding corrodes and the waste package fails.[14] If we choose 10 percent as an example, no more than 3 out of 30,000 waste packages can fail per year for 10,000 years. Otherwise, releases of C-14 dioxide would violate the limits in EPA and NRC regulations.

Currently the EPA and NRC regulations are forcing the DOE to design a waste package that completely contains the carbon-14 for 10,000 years with essentially no margin for failure (i.e. design a 10,000 year waste package). Preferably, the DOE could persuade the EPA and the NRC to start rulemakings that would exempt carbon-14 dioxide from the release requirements. The exemption would not measurably increase risks to public health and safety, and would reduce costs significantly.

A 10,000 year waste package may increase public confidence or compensate for geologic uncertainties, but it should not be developed solely to control releases of carbon-14 dioxide. The benefits do not justify the cost. We estimate that a 10,000 year waste package - as opposed to a 1,000 year package - would reduce carbon-14 releases only 10 Curies/year and reduce the corresponding exposure only 0.0005 mrem/yr [15]. These negligible benefits to public health and safety could add $2.4 billion to the cost of the waste packages.

The current reference thin-wall waste packages will cost roughly between $45,000 and $80,000 each, depending on the vendor, material selected, and fabrication and closure methods. If 30,000 packages are needed, the total cost would be $1.35 billion to $2.4 billion. More conservative designs, such as thick-walled packages and multi-barrier packages, with either metallic or ceramic inserts may double these costs. The fabrication of these more conservative packages will need development, particularly those made of ceramic materials; and even then, successful compliance with NRC and EPA requirements is not assured.

It could be argued that the DOE should design a 10,000 to 1,000,000 year waste package regardless of what the regulations say. For example, some scientists have suggested that the United States pattern its repository program after Sweden's and design a waste package that will last 1 million years. Such a package would increase public confidence and partially compensate for geologic uncertainties. Yet, apart from costs, there are disadvantages, namely, the U.S. regulations.

The U.S. repository is constrained by regulations that do not exist in Sweden. These regulations demand proof (i.e. reasonable assurance) and verification that the waste package will survive a multitude of small-probability events; possibly as low as one chance in 2 million. Thus, even if the United States built a waste package that was as durable as Sweden's, we could probably never prove, to the satisfaction of our regulations, that it would last 1 million or even 10,000 years.

In its proposed amendments to 40 CFR Part 61[16] the EPA suggests several options for limiting radioactive contamination to the air. The limits apply to a variety of industries, some nuclear some not. Among the suggestions, proposed limits for risk, 1×10^{-6} (the annual probability of contracting a fatal cancer over a 70-year lifetime), and dose, 0.03 mrem/yr, are the most restrictive and could force some uranium mills out of business. The EPA states,

> Under Approach D, no new mill tailings may be produced starting after the effective date of the rule [40 CFR Part 61]. EPA is forced to go to this extreme solution because it knows of no way to manage new tailings that will result in risks of less than 1×10^{-6}. (54FR9646).

The amendments to 40 CFR Part 61 also state that gaseous emissions from a repository are so low that the EPA will not regulate them. Yet they are regulated, albeit unintentionally, in another EPA regulation, 40 CFR Part 191, which applies just to repositories. Here the EPA imposes on gaseous emissions (mainly carbon-14 dioxide) limits that are much more extreme than the most conservative limits proposed in 40 CFR Part 61. A repository should be governed by more equitable limits, particularly when repository risks are generally orders of magnitude less than the radioactive risks posed by other industries.

A repository filled with spent fuel from 70,000 metric tons (MT) of heavy metal contains about 100,000 Curies of carbon-14. Currently, 40 CFR Part 191 limits carbon-14 releases to 100 Curies/1,000 metric tons of heavy metal.[1] For a repository filled with 70,000 metric tons, carbon-14 releases can average no more than .7 Curies per year for 10,000 years. If released as carbon dioxide, the .7 Curies of carbon-14 corresponds to a maximum dose of about

0.00003 mrem/yr which is three orders of magnitude lower than most restrictive limits proposed for other industries (The risk proposed in 40 CFR Part 61, 1 x 10^{-6}, corresponds to annual dose of about 0.03 mrem).

The rate at which carbon-14 leaves the waste package also affects compliance with the NRC requirement, 10 CFR Part 60.113(a)(ii)(B):

> The release rate of any radionuclide from the engineered barrier system [in the case of Yucca Mountain, from the waste package] following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following the permanent closure...[2]

By comparison, C-14 produced in the coolant water during the reactor operation can be discharged without any regulatory constraints. Compared to the 1,000 to 5,000 Curies of rapid release fraction of C-14 that may be released from a repository in 10,000 years, the same 70,000 MTHM within reactors would have discharged 20,000 curies of C-14 directly into the atmosphere in less than 100 years without violating any existing EPA or NRC regulations. Furthermore, a reprocessing plant could release the entire inventory remaining in the spent fuel, i.e., 100,000 Curies, into the atmosphere without violating any regulation. The United States is not reprocessing any commercial spent fuel at present; however, it is not forbidden.

A typical 1500 MTHM/yr fuel reprocessing plant, if no C-14 control is practiced (currently not required), would release 860 Curies per year (Ci/yr) during its operation without violating any regulations.[17] In processing defense spent fuel, the Savannah River Plant (SRP) reported a total production of 392 $\pm$ 40 Ci/yr[18]. In 1986, SRP reported a gaseous release of 56 curies.[8]

The irony of this situation is that C-14 within spent fuel may be freely released from a variety of nuclear facilities, but the moment the spent fuel is placed in an underground repository, excessive limits apply. The DOE could circumvent these limits if, prior to emplacement into the repository, C-14 were removed from spent fuel exterior surface and discharged into the atmosphere. This discharge would be acceptable under the current EPA and NRC regulations, but poses a greater threat to public health and safety than an underground discharge.

By emplacing the waste underground and distributing the release in 10,000 years, the absolute total infinite time dose commitment is reduced to approximately one half by the radioactive decay and the dose commitment to any given human generation is reduced by two orders of magnitude. Therefore, in order to circumvent EPA and NRC requirements, the DOE could spend additional money to remove C-14 from spent fuel but increase, without violating any regulations, the dose commitment to the present and future generations.

According to the NRC requirement, carbon-14 releases cannot exceed 1 Curie per year for 10,000 years (unlike the EPA limit, .7 Curies/year, this is not an average yearly release, but a maximum release for any year). Assuming that 1,000 Curies of the carbon-14 inventory will oxidize to carbon-14 dioxide (the rapid release fraction), the DOE has assessed that the NRC and perhaps the EPA requirements will be violated if more than 30 out of 30,000 waste packages fail in any year for 10,000 years.[4,11]

More recent data, however, indicate that the rapid release fraction could be as high as 5,000 Curies and some German data suggest 10,000 Curies. As the rapid release fraction increases, fewer packages can fail without violating the EPA and NRC requirements. For example, if the fraction is 5,000 Curies, then no more than 6 packages can fail per year. If, however, the rapid release fraction is 10,000 Curies, fewer than 3 out of 30,000 packages can fail in any year for 10,000 years. A failure rate this low would drive the DOE to design a more conservative waste package that could double the costs but reduce an individual's exposure by a few ten thousandths of a mrem/yr.

For the sake of an example, we will take the highest suggested fraction of Carbon-14 that can be rapidly released, 10,000 Curies, and a realistic failure rate for the waste packages, 30 failures/year,[6] and examine the impacts of violating the EPA and the NRC requirements by an order of magnitude. If thirty waste packages failed every year, after 1,000 years all 30,000 packages would have failed and 10,000 Curies of carbon-14 dioxide would eventually be released into the atmosphere; an average of 10 Curies/year. Even if the gas escaped directly into the atmosphere (assuming no retention in the host rock) the carbon-14 dioxide poses little threat to the public's health and safety.

The atmosphere already contains 4.4 million Curies of carbon-14, about 2 percent of the total global inventory of 220 million curies. Moreover, the atmosphere plus other natural sources produce carbon-14 at the rate of 26,700 Curies/year. In comparison, 10,000 Curies of carbon-14 dioxide (the highest fraction of a repository's inventory that can be rapidly released) would increase the atmospheric inventory by 0.23%. If the repository released carbon-14 at a rate of 10 Curies/year (which is ten times higher than the EPA and NRC limits) it would increase carbon-14 production by .04%. These miniscule increases can hardly threaten the world's population, but would expose some hypothetical individual who lives, eats and sleeps on top of a repository to a very small incremental dose of 0.0005 mrem/yr (which is likewise ten times higher than what the EPA and NRC limits infer, because in the case of carbon-14, the relationship between Curies and dose is linear).

Natural sources of carbon-14 expose individuals to much higher doses. For example, carbon-14 in the human body in equilibrium with atmospheric carbon-14 exposes one to 1.3 mrem/yr.[19]

Other sources of radiation expose individuals to even higher doses. For example, a chest X-ray exposes one to 20 to 70 mrem and the natural background radiation (at sea level) exposes one to 100 mrem/yr. Our point is that reducing radiation exposures by 0.0005 mrem/yr is a poor reason, in itself, to develop and manufacture a 10,000 year waste package. As stated earlier, a 10,000 year package could increase public confidence and compensate for geologic uncertainties, but it should not be constructed just to contain carbon-14 dioxide. The benefits simply do not justify the expense.

When the EPA and NRC first wrote their regulations, they doubted that their release requirements would apply to gasses.[20,21] At the time the regulations were written, saturated sites were the leading contenders for a repository. Both agencies wrote limits believing that solid radionuclides (the NRC forbids emplacing liquid waste) would dissolve within a saturated repository and reach the accessible environment via a groundwater pathway (the EPA scenario for human intrusion is an exception). Neither assumed that radioactive gases within an unsaturated repository would rise to the surface and never contact the groundwater. Consequently, the EPA and the NRC unintentionally included release limits for radioactive gases, and now the DOE is trying to comply with them.

Two years after 10 CFR Part 60 was issued, the NRC amended it so that it would also apply to disposal within the unsaturated zone. At that time, the NRC foresaw that an unsaturated repository (i.e., a repository excavated above the water table) could challenge the relevance of their controlled release requirement. The Commission stated,

> Vapor transport has been identified by the Commission's staff as potential concern associated with HLW disposal in the unsaturated zone (49FR5934, February 16, 1984).

The Commission knew that its requirement neglected vapor transport: a phenomenon peculiar to the unsaturated zone. After a waste package fails, contaminated vapor, such as carbon-14 dioxide, would escape through the opening and rise through the unsaturated rock. The Commission could not think of ways to tailor its regulations to this phenomenon. So they simply postponed a decision:

> Therefore the Commission views vapor transport as another issue which must be evaluated on a case-by-case basis to determine its effects on a particular site (Ibid).

Fortunately, there is a provision, 10 CFR Part 60.113(b), that allows the Commission, on a case-by-case basis, to approve some other radionuclide release rate. The provision states,

> On a case-by-case basis, the Commission may approve or specify some other radionuclide release rate...provided that the overall system performance objective, [the EPA Standard, 40 CFR Part 191] as it relates to anticipated processes and events, is satisfied.

We believe that the Commission had this provision in mind when it stated that vapor transport is an issue that must be evaluated on a case-by-case basis. Vapor transport is a phenomenon peculiar to the unsaturated zone, but the controlled release requirement was written for disposal within the saturated zone. Since the phenomenon does not fit the requirement, the DOE has valid grounds to take advantage of 10 CFR Part 60.113(b) and petition the NRC to exempt radioactive gases from its release rate requirement.

RECOMMENDATIONS

The EPA should correct the discrepancies between its repository regulations, 40 CFR Part 191 and its industry regulations, 40 CFR Part 61. The regulations of 40 CFR Part 191 become excessively and needlessly conservative when applied to radioactive gases, particularly carbon-14 dioxide. The EPA should regulate repository and industrial emissions in one regulation, 40 CFR Part 61, or exempt carbon-14 dioxide from all its regulations. Since the EPA is currently revising 40 CFR Part 191, this would be good time to make these corrections.

The NRC should allow a higher release rate for carbon-14 dioxide or simply exempt it. The NRC regulations of 10 CFR Part 60 are driving the DOE to develop a 10,000-year waste package simply for the sake of controlling carbon-14 dioxide. In regulating nuclear power plants, the NRC advocates radiation doses to the public that are as low as reasonably achievable (ALARA). If the ALARA principle were applied to a repository, it would not be reasonable to reduce carbon-14 doses by 0.0005 mrem when the only way of achieving this reduction is to contain the waste in a 10,000-year waste package. As stated in the title of this paper, the negligible benefits do not justify the high costs.

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